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CALCULATION OF THE COHESIVE ENERGY OF METALS BY THE FERMI-THOMAS METHOD

/The following is a full translation of an article written by Ch'eng K'ai-chia, appearing in Wu-li Hsueh-pao (Journal of Physics), Peiping, Vol. XIV, No. 2, 1958, pages 106-112./

Abstract

The Fermi-Thomas method is used in this paper to calculate the cohesive energy of metals. The preliminary results obtained indicate that there is a minimum value involved in the relation between the energy and the crystal lattice constant of a metal. For heavy metals, the atomic spacings corresponding to this minimum value are shown to be close to the experimental values; however, for univalent alkali metals, the theoretical values are too small, and the cohesive energies so obtained are much larger than the experimental values. The author tends to think that such discrepancies are due to errors typical in an application of the Fermi-Thomas method to atoms, and that they are not particularly pertinent to the application to metals.

I. Introduction

Fermi statistics was first applied to atomic problems by Thomas[1] in 1926. It is assumed in this theory that for all values of momentum smaller than a fixed p, the distribution of electrons is uniform in phase space, i.e., in each element in phase space of volume h^3 there are two electrons; further, it is assumed that there exists a potential energy V in the atom, which depends only on the distance r from the nucleus, and thus by means of the Poisson equation one can construct

the proper equation for the potential energy in the atom. Practical calculations of the cohesive energies of atoms were carried out by Milne [2] and by Baker[3] ; their results are greater than that obtained by Hartree's method by 20 percent. Corrections for boundary effects and exchange energy made by Scott[4] bring the calculations into good agreement with those using Hartree's method and with the experimental data; the error involved is less than 3 percent. The success of the application to atoms has made possible the application to metals. Since in the application of the method of Wigner and Seitz[5] to calculate the cohesive energies of metals, it is possible to get agreement with experimental data only for alkali metals, and the application to multivalent metals is very difficult, one would think that the statistical method could be used to overcome such difficulties. The work by Slater and Krutter[6] in 1935 showed little prospect of obtaining the cohesion of metals; no minimum value occured in their curve of the total energy of the metal versus the atomic spacing; the results were not significantly improved even after correction for exchange energy. Such results, according to them, were due to the omission of the correlation energy. A more systematic study begun in 1936 by Gombas[7] shows that the problem in the calculations of Slater et al. lies not in the omission of the correlation energy but in the unjustified treatment in their statistics of attributing the same properties to the valence electron and the electrons in the ions. According to Gombas, one could get results in agreement with experimental data and the cohesion of the metallic lattice points if one regarded the valence electrons as free electrons distributed uniformly in metals and having different statistics from that of the electrons in the ions, and took into account the cohesive energy and correlation energy of free electrons, the mutual interactions of ions, together with the exchange energy of the valence electrons and the electron shells of the ions, etc. Only alkali and alkali-earth metals are considered in Gombas calculations; the valence electrons of such metals are practically all free electrons, and so the results are close to those obtained by Hartree's method. However, for other metals such an application would have fundamental difficulties due to the lack of clear distinction between valence electrons and the electrons in the ions. One finds, for instance, in the application of the Hartree wave function to copper, that for the 10 e electronic charge of the d shell about

0.4 e is distributed outside the atomic sphere having a volume equivalent to that of one lattice unit. In other words, the d shells expand into comparatively broad energy bands in crystals, and the d electrons would tend to play a role almost as important as that of the valence electrons[8] as far as cohesive energy is concerned. There is no sufficient reason to make a sharp distinction between valence electrons and inner-shell electrons. Further, results relevant to practical conditions can be obtained with statistical methods only when the system under consideration has a large number of particles; and it is hardly meaningful to proceed statistically with only a handful of particles. After the separation, made by Gombas, of the valence electrons and inner-shell electrons into different categories, it is hard to understand why the valence electrons as individual particle systems could obey statistical laws. The results of Gombas calculation, though achieving some quantitative agreement with experiments, could likely be a sort of accidental coincidence, void of firm foundation.

In this paper, we hope to find a general method of solving the problems of metals of many valence electrons, or rare-earth metals. We believe that the limitation in the work of Slater and Krutter is due neither to the omission of correlation effects nor to the combined statistics of valence electrons and inner-shell electrons, but that it arises from the improper boundary conditions they employed. In the following, we will try to set up proper boundary conditions to solve the Fermi-Thomas equation, and obtain better results.

II. The Fermi-Thomas Equation and Its Boundary Conditions

In the application of the sphere approximation method of Wigner and Seitz, we set up a sphere of radius R around every atom, determined by the condition that the volume of the sphere be equal to the atomic volume and that every sphere be electrically neutral, with the mutual interaction between spheres neglected in the calculations. We need only to calculate the energy of each sphere. From this energy we subtract the energy of an isolated atom and obtain the cohesive energy.

Suppose in every atomic sphere we can define a potential function $V(r)$ depending on the distance r from the nucleus alone; then $-eV(r)$ is just the sum of the potential energies of an electron due to its interaction

with the nucleus and with other electrons. The kinetic energy of the electron is $p^2/2m$ and its total energy $-eE$ is given simply by

$$\frac{p^2}{2m} - eV = -eE$$

In the classical case the momentum p must be a real number, i.e., $-e(E-V)$ must be a positive number, in order that the motion be well defined. The total energy is $-eE_0$ corresponding to the maximum momentum P of the electron. Thus $-eE_0$ is just the energy of an electron on the Fermi surface in metals. From the above formula,

$$P = [-2me\ (E_0 - V)]^{\frac{1}{2}}\ .$$

According to Fermi statistics, the space charge density is

$$\rho_e = -\frac{8\pi e P^3}{3h^3} = \left(-\frac{8\pi e}{3h^3}\right)[-2me(E_0 - V)]^{\frac{3}{2}}.$$

Substituting in the Poisson equation,

$$\nabla^2 V = -4\pi\rho_e.$$

with the assumption of spherical symmetry of the electron distribution, we obtain

$$\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial V}{\partial r}\right) = \frac{32}{3}\pi^2\frac{e}{h^3}[2me(V - E_0)]^{\frac{3}{2}}. \qquad (1)$$

Set $r = ux$, with

$$\mu = a_0\left(\frac{9\pi^2}{128z}\right)^{\frac{1}{3}} = \frac{0.4685}{z^{\frac{1}{3}}}\ \text{Å}, \qquad (2)$$

$$V - E_0 = \frac{\gamma\phi}{x}, \quad c\gamma = \frac{ze^2}{\mu} = 2.2590\, z^{\frac{4}{3}}\ \text{ry}. \qquad (3)$$

It is seen from the above formulas that

$$Ze\phi = (V - Eo)r, \text{ as } r \to 0,\ r(V-Eo) \to Ze,$$

and thus,

$$\phi\ (0) = 1. \qquad (4)$$

With the new variable, we can write the Poisson equation as

$$\phi'' = \frac{\phi^{\frac{3}{2}}}{x^{\frac{1}{2}}}. \qquad (5)$$

According to Slater and Krutter, the condition that the atomic sphere be neutral simply implies that at the boundary the electric field vanishes. Thus,

$$\left.\frac{\partial V}{\partial r}\right|_{r=R} = 0,$$

where R is the radius of the sphere. That is,

$$\phi'_X = \frac{\phi_X}{X}, \quad X = \frac{R}{\mu}. \tag{6}$$

Here ϕ_X is the value of ϕ at the boundary of the sphere. With the boundary condition (4), one can then proceed to solve Eq. (5) numerically. Given an initial slope ϕ_0, there corresponds a graph of a solution ϕ versus x. From condition (6) it is clear that the tangent to the curve made from the origin will meet the curve at the point whose abscissa corresponds to the radius of the atomic sphere. For a given initial slope, the solution for an isolated atom is given by ϕ whose graph against x is asymptotically tangent to the x-axis at infinity. This numerical solution provides us a sequence of values of X, ϕ'_0, ϕ'_X and ϕ_X. From these values one can then calculate the potential energy, kinetic energy and total energy of the crystal.

The total potential energy of an electron in the atom is

$$\frac{1}{2}\int_0^X \rho_e V dv + \frac{1}{2} zeV_0,$$

Pe being the electron density, and Vo being the electric potential due to the electron at the position of the nucleus.

$$\rho_e = -\frac{Z}{4\pi\mu^2}\frac{\phi^{\frac{3}{2}}}{x^{\frac{3}{2}}}, \quad V_0 = \int_0^X \frac{\rho_e}{r} dv.$$

After substitution, with help of $\phi'' = \phi^{\frac{3}{2}}/x^{\frac{1}{2}}$, one can perform an integration by parts similar to that of Milne, and obtain

$$\text{Potential energy} = \frac{1}{2}\int_0^X \left(-\frac{Z}{\mu^2}\frac{\phi^{\frac{3}{2}}}{x^{\frac{3}{2}}}\right)\left(\frac{Z\phi}{x} + E_0\right)\mu^3 x^2 dx + \frac{1}{2} ze\int_0^X (-Z)\frac{\phi^{\frac{3}{2}}}{x^{\frac{1}{2}}} dx =$$

$$= -\frac{1}{2} ze\gamma \int_0^X \frac{\phi^{\frac{3}{2}}}{x^{\frac{1}{2}}} dx - \frac{1}{2} zeE_0 \int_0^X \phi^{\frac{3}{2}} x^{\frac{1}{2}} dx - \frac{1}{2} ze\gamma \int_0^X \frac{\phi^{\frac{3}{2}}}{x^{\frac{1}{2}}} dx =$$

$$= -\frac{1}{2} ze\gamma \left[\frac{5}{7} \left(\phi\phi' - x(\phi')^2 + \frac{4}{5} x^{\frac{1}{2}} \phi^{\frac{5}{2}} \right) \right]_0^X -$$

$$-\frac{1}{2} zeE_0 \left[x\phi' - \phi \right]_0^X - \frac{1}{2} ze\gamma \left[\phi' \right]_0^X.$$

From the boundary condition ϕ (0) = 1, one can obtain

$$\text{Potential energy} = \frac{3}{7} ze\gamma \left[2\phi_0' - \frac{2}{3} X^{\frac{1}{2}} \phi_X^{\frac{5}{2}} \right] - \frac{1}{2} zeE_0 - \frac{1}{2} ze\gamma\phi_X'. \qquad (7)$$

The maximum kinetic energy of the electron is $e(V - E_0)$. The mean kinetic energy is 3/5 this value, and so according to Fermi statistics the kinetic energy is given by

$$\text{Kinetic energy} = \int_0^X \frac{3}{5} (V - E_0)\rho_e dv = \frac{3}{5} ze\gamma \int_0^X \frac{\phi}{x} \frac{\phi^{\frac{3}{2}}}{x^{\frac{3}{2}}} x^2 dx =$$

$$= \frac{3}{7} ze\gamma \left[-\phi_0' + \frac{4}{5} X^{\frac{1}{2}} \phi_X^{\frac{5}{2}} \right]. \qquad (8)$$

Total energy = potential energy + kinetic energy

$$= \frac{3}{7} ze\gamma \left[\phi_0' + \frac{2}{15} X^{\frac{1}{2}} \phi_X^{\frac{5}{2}} \right] - \frac{1}{2} zeE_0 - \frac{1}{2} ze\gamma\phi_X'. \qquad (9)$$

Now the remaining problem is to determine Eo. The condition used by Slater and Krutter to determine Eo is as follows: the electric potential in the close vicinity of the nucleus $(r \to 0)$ is not affected by the interatomic spacing, or, the electric potential in the close vicinity of a nucleus in a metal should be equivalent to that corresponding to a nucleus in an isolated atom. Thus we obtain

$$E_0 = \gamma[\phi_0'(\infty) - \phi_0'(X)].$$

Here, $\phi_0'(\infty)$ is the initial slope corresponding to the solution for an isolated atom; and

$$V_0 = \int_0^\infty \frac{\rho_e}{r} dv = -\gamma\phi_0'(\infty).$$

It follows that

$$\text{Total energy} = \frac{3}{7} ze\gamma \left[\phi_0' + \frac{2}{15} X^{\frac{1}{2}} \phi_X^{\frac{3}{2}}\right]. \tag{10}$$

Starting with this formula[1)] one fails to obtain a minimum value for the curve of the total energy against X; in other words, from the above one is not able to illustrate the stability of the cohesion of metals.

In our opinion the above boundary condition is not adequate. The electric potential of an electron is the near neighborhood of the nucleus is the work done in taking an electron from infinity to that neighborhood. This should obviously be different for an isolated atom and for an atom in a metal. To bring an electron from infinity to the inside of a metal one has to overcome the boundary effect; in other words, the actual work done is different from that for an isolated atom by the work function of the metal. Slater et al. overlooked this point, and so could not obtain a stable metallic cohesion.

After some calculations, we have reached the conclusion that their assumption together with the assumption of zero potential at the boundary of the sphere is exactly equivalent to the following assumption:

$$V_0 = \int_0^X \frac{\rho_e}{r} dv = -\gamma(\phi_X' - \phi_0')$$

the latter assumption leads to results identical to Eq. (10). However, this latter assumption was taken by Slater et al. as being of little physical meaning.

Fermi-Thomas statistics takes into consideration all the electrons in its calculation of electric charge density at a point in space. When we calculate the electron potential energy from this charge density, we should notice that the potential energy is the energy of the electron due to the potential field of the nucleus and other electrons. Therefore, rigorously speaking, to find the potential energy of a given electron, one should proceed with a statistics that considers the potential energy of all electrons except for the given one. However, to find the electric charge distribution one

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1) (10) seems to involve some error. Since in Slater's paper the actual calculations are not fully presented, we cannot say whether Slater's calculation is accurate or not.

has to consider the effect of this particular electron on all other electrons. In this manner, the potential energy of the given electron can be found from the electric charge density distribution. Since our statistical object is a large system of Z electrons, the density distribution of Z - 1 electrons can be substituted by that of Z electrons; the resulting fractional error in the substitution is not larger than $\frac{1}{Z}$. But in considering the electric potential of a given electron at the boundary of the atomic sphere due to all other electrons, one should take $\frac{e}{R}$ and not zero. Since the potential seen by the given electron is due to Z - 1 negative and Z positive charges, the electric potential at the boundary cannot be regarded as vanishing as in the case of a potential of an averagely neutral sphere.

The boundary condition is thus:

$$V|_{r=R} = \frac{e}{R} = \frac{e}{\mu X} = \gamma \frac{1}{zX},$$

So, $$E_0 = V_R - \gamma \frac{\phi_X}{X} = \gamma \frac{1}{zX} - \gamma \phi'_X.$$

After substitution in Eq. (9) and some manipulation we obtain

$$\text{Total energy} = \frac{3}{7} ze\gamma \left[\phi'_0 + \frac{2}{15} X^{\frac{1}{2}} \phi^{\frac{3}{2}} - \frac{7}{6} \frac{1}{zX}\right]. \qquad (11)$$

III. Results of the Calculation of the Cohesive Energy

The cohesive energy of a metal is the difference between the total energy of an atom in the metal and the total energy of the isolated atom. The order of magnitude of the cohesive energy is about 10^{-4} to 10^{-6} that of the total energy of an isolated atom. Therefore, in order to calculate accurately the cohesive energy one has to solve Eq. (5) accurately so as to get about eight significant figures. So far there is no such accurate numerical solution. In the following we will employ two methods of calculation and compare the results. These two methods are basically the same. We shall first eliminate the energy of the isolated atom by an approximate expansion, and then correct our calculation with help of the numerical solution of Eq. (5).

1. Approximate expansion

Set $\phi = \phi_A + \psi.$

Here ϕ_A is the function for the isolated atom, is an additional function. They are of the same order of magnitude for large r. However, we take $\phi_A > \psi$ in general and obtain the approximate solution by substituting the above relation in Eq. (5):

$$\phi_A'' + \psi'' = \frac{(\phi_A + \psi)^{\frac{3}{2}}}{x^{\frac{1}{2}}} \simeq \frac{\phi_A^{\frac{3}{2}}}{x^{\frac{1}{2}}} + \frac{3}{2}\left(\frac{\phi_A}{x}\right)^{\frac{1}{2}} \psi.$$

In the isolated atom, ϕ_A satisfies the equation:

$$\phi_A'' = \frac{\phi_A^{\frac{3}{2}}}{x^{\frac{1}{2}}}, \tag{12}$$

and hence,

$$\psi'' = \frac{3}{2}\left(\frac{\phi_A}{x}\right)^{\frac{1}{2}} \psi. \tag{13}$$

One can make use of the solution to Eq. (12) by Bush and Caldwell[9] to solve Eq. (13) numerically. Equation (13) is a linear differential equation, and so its solution may be different by an arbitrary constant B which can be determined by boundary conditions. From the boundary condition,

$$\phi_X' = \frac{\phi_X}{X}, \quad \phi_A' + B\psi'|_{x=X} = \frac{\phi_{A_X} + B\psi_X}{X}$$

we obtain

$$B = \frac{\phi_{A_X} - X\phi_{A_X}'}{X\psi_X' - \psi_X}. \tag{14}$$

B is a function of the radius X of the atomic sphere. On substituting the above expressions in Eq. (11) and eliminating the function for isolated atom, we obtain the cohesive energy ΔE in terms of the atomic radius X:

$$\Delta E = \frac{3}{7} ze\gamma \left[B\psi_0' + \frac{2}{15} X^{\frac{1}{2}} (\phi_{A_X} + B\psi_X)^{\frac{5}{2}} - \frac{7}{6}\frac{1}{zX}\right].$$

There is a minimum value in the graph of ΔE against X. The radii of the atomic spheres for several metals, corresponding to this minimum value, are shown in table 1.

Table 1 (R in units of Å)

Element	Na	Al	K	V	Cr	Co	Cu	Mo	Ag	Ta	Pt
Atomic number	11	13	19	23	24	27	29	42	47	73	78
R Experiment	2.08	1.62	2.57	1.47	1.41	1.39	1.41	1.54	1.59	1.61	1.54
R Theory	1.48	1.49	1.44	1.39	1.39	1.35	1.38	1.36	1.32	1.26	1.23

The values of ΔE corresponding to the minimum value are all too large by 1 or 2 orders of magnitude. This is due to the approximation in the calculation for the isolated atom. From the nature of the approximation one can roughly estimate such errors.

2. Numerical evaluation

The numerical integeration of Eq. (5) has been carried out independently by Slater and Krutter, by Feynman, Metropolis and Teller[10], by March[11] and by Latter[12], etc. Among these the calculation by Latter is most exhaustive. However, there are only five significant figures in his tabulated data; and such are not sufficient for an evaluation of the cohesive energy. Here we have utilized his tables to find the relation between ϕ_o, ϕ_x and X, while we estimate two more figures. The numerical results are as shown in Table 2. The initial slope is about - 1.588073 for the isolated atom solution.

Table 2

X	$-\phi_0'$	ϕ_X	X	$-\phi_0'$	ϕ_X
4.905	1.583768	0.20099	8.733	1.587854	0.071490
5.229	1.584923	0.18058	9.565	1.587932	0.059868
5.401	1.585302	0.17095	9.858	1.587970	0.056426
5.506	1.585504	0.16511	10.804	1.588023	0.046934
5.850	1.586275	0.14899	11.300	1.588030	0.042832
6.177	1.586675	0.13547	11.963	1.588033	0.038086
6.206	1.586687	0.13433	12.369	1.588036	0.035529
7.014	1.587314	0.10539	13.771	1.588040	0.028322
7.385	1.587435	0.098098	15.471	1.588045	0.022021
7.790	1.587560	0.088823	15.870	1.588050	0.020825
8.015	1.587643	0.084193	16.00	1.588060	0.020454
8.588	1.587798	0.073821			

If we substitute in Eq. (11) the numbers given in Table 2, we can obtain a graph of ΔE against X; this graph has a minimum value which corresponds to atomic radii as shown in Table 3.

Table 3 (R in units of Å)

Element	Na	Al	K	V	Cr	Co	Cu	Mo	Ag
Atomic number	11	13	19	23	24	27	29	42	47
R Experiment	2.08	1.62	2.57	1.47	1.41	1.39	1.41	1.54	1.59
R Theory	1.47	1.49	1.41	1.37	1.38	1.37	1.40	1.52	1.57

The ΔE values obtained are equivalent to those obtained by the previous approximation method.

IV. Discussion

From the above tables we see that the atomic radii derived from the theory are basically in agreement with the experiments. For multivalent and B-group metals, e.g., V, Cr, Co, Cu, Ag, etc., the agreement is very good, but for A-group univalent alkali metals, Na, K, etc., the deviation becomes larger. Since the cohesive energy of

alkali metals is due chiefly to the change in electron cloud distribution of one single valence electron, the reason for the deviation is easily seen to be the large error caused by the application of Fermi statistics to the single-electron system. Therefore one should not expect consistent results for univalent alkali metals.

As to the great deviation between the theoretical and experimental values of cohesive energy. We would attribute it to the following reason: the cohesive energy of metals is of the order of 10^{-4} to 10^{-6} that of the total electronic binding energy of the atom; a 1 percent or even 0.1 percent error in the total energy of the atom is enough to cause a completely erroneous result in the cohesive energy. Since the Fermi-Thomas method is accurate to the order of $\frac{1}{Z}$, it would be hard to do any accurate numerical calculation of the cohesive energy. If one can find a mathematical technique for cancelling the energy of the atom from the final result so that it will not appear in the final result for the cohesive energy, then one can eliminate a great portion of the error.

In the Fermi-Thomas statistical method, we use the classical concept that the electron momentum p has to be real so that the electron kinetic energy will be positive. This, however, is not consistent with the quantum-mechanical concept. Quantum mechanics indicates that in the outer part of the atom, the kineticenergy of the electron can be negative. When the Fermi-Thomas method is applied to the calculation of the total energy of the atom, no significant error could occur from omitting this negative part of the kinetic energy; however, the numerical value of the cohesive energy could be affected immensely. Therefore, in an accurate calculation, corrections should be made according to an estimate of magnitude of this negative kinetic energy. In actual problems, for example, the calculation of energy of an alloy, one can make use of the experimental data for pure metals to decide on this part of the correction.

This paper presents a preliminary calculation of the cohesive energy of pure metals without taking into consideration the exchange energy and the correlation energy. Nevertheless, the results obtained prove to be an improvement over the work by Slater and Krutter, since the negative minimum value for the cohesive energy of metals and the radii of the atomic spheres obtained are basically in agreement with the experimental data. It is hoped that, after some modification of the above method, it would be possible to calculate the cohesive energy of heavy metals, e.g., the rare-earth metal alloys

and the black metal alloys. It is therefore of considerable significance for the study of alloys.

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